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14. ABSTRACT
The recent discovery of a two-dimensional electron gas (2DEG) at the interface between insulating perovskite oxides $SrTiO_3$ and $LaAlO_3$ was made possible by advances in atomic layer controlled growth. These advances have led to the creation of atomically-abrupt interfaces between novel complex oxide materials. It has been demonstrated that the conducting layer can be localized within a few nm of the interface, and that the carrier concentration can be altered with an electric field and/or lattice strain. We have created a strong interdisciplinary collaboration with the expertise in US and Korea required to attack the fundamental issues in this exciting, emerging field. We have for the first time directly imaged **the charge carrier densities and spatial distributions** at the (001) $LaAlO_3/SrTiO_3$ heterointerfaces by *in-situ* TEM holography. The new understanding from this measurement will ultimately lead to the nm-scale writing of *up* and *down* polarization ferroelectric domains that permit the design of nonvolatile switchable devices. We envision logic devices and tunable metamaterials with switchable electron current based on 2D interface materials.

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Final Report for AOARD Grant FA2386-13-1-4136

"Direct Imaging of Charge Density Modulation in Switchable Two-Dimensional Electron Gas at the Oxide Hetero-Interfaces by using Electron Beam Inline Holography"

August 16, 2015

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Abstract

The recent discovery of a two-dimensional electron gas (2DEG) at the interface between insulating perovskite oxides $SrTiO_3$ and $LaAlO_3$ was made possible by advances in atomic layer controlled growth. These advances have led to the creation of atomically-abrupt interfaces between novel complex oxide materials. It has been demonstrated that the conducting layer can be localized within a few nm of the interface, and that the carrier concentration can be altered with an electric field and/or lattice strain. We have created a strong interdisciplinary collaboration with the expertise in US and Korea required to attack the fundamental issues in this exciting, emerging field. We have for the first time directly imaged **the charge carrier densities and spatial distributions** at the (001) $LaAlO_3/SrTiO_3$ heterointerfaces by *in-situ* TEM holography. The new understanding from this measurement will ultimately lead to the nm-scale writing of *up* and *down* polarization ferroelectric domains that permit the design of nonvolatile switchable devices. We envision logic devices and tunable metamaterials with switchable electron current based on 2D interface materials.

Introduction

This project is a collaborative effort to explore the fundamental scientific issues of the growth and novel properties of oxide hetero-interfaces. Specific tasks are **(1) atomic layer epitaxial growth** and **characterization** of switchable two-dimensional oxide hetero-interface materials; (**2) direct imaging of charge carrier densities by inline holography and electrical transport** of 2DEG oxide hetero-interfaces. Our goal is to achieve an atomic-level understanding of the growth and characteristics of oxide hetero-interfaces, with advanced properties and new functionalities.

Experiment:

Atomic Layer Controlled Growth of Oxide Hetero-Interfaces

We have grown $LaAlO_3/SrTiO_3$ oxide hetero-interfaces by using pulsed laser deposition atomic with *in-situ* reflection high-energy electron diffraction (RHEED). We have incorporated a differentially pumped high-pressure RHEED providing essential real-time monitoring and feedback to the growth process, and provide atomic-layer control of epitaxial oxide heterostructures at high oxygen partial pressure.

Direct imaging of 2DEGs: Inline electron holography

Electron holography in a transmission electron microscope (TEM) is the unique tool capable of mapping electrostatic potentials and associated charge distributions in semiconductor materials, and therefore regarded as the only tool that can completely visualize the spatial distribution of the 2DEG in oxide hetero-interfaces. Compared to the classical off-axis technique, recently developed inline holography is dramatically more powerful, as it can directly reconstruct the phase information of electron wavefunction transmitted through the sample from a series of bright-field (BF) images recorded at different planes of focus (Fig. 1).

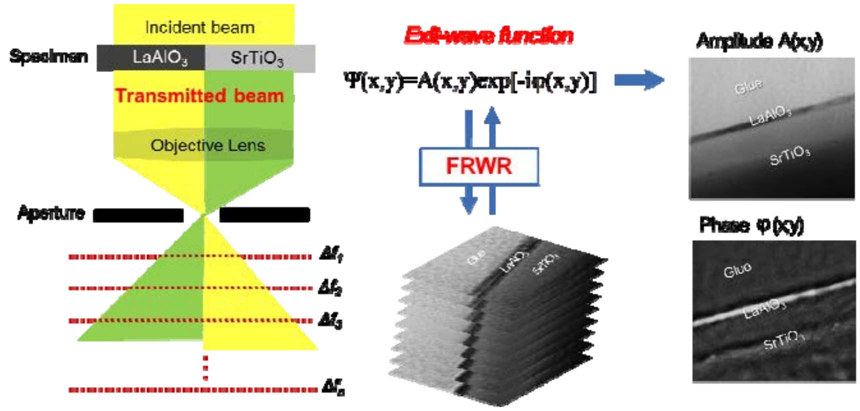

Figure 1. Schematics illustrating inline electron holography to retrieve the phase and amplitude of exit wavefunction. A through-focal series of TEM images is recorded at different focus planes. The phase of exit-wave, φ(x, y), is reconstructed using full resolution wave reconstruction (FRWR) algorithm developed by Koch[5]

Assuming that TEM sample behaves as a phase object, the obtained phase map can be converted to the electrostatic potential distribution with a straightforward linearization of the phase of the exit wavefunction. The local electrostatic potential generated by 2DEG obeys Poisson's equation

$$\rho = -\varepsilon_0 \varepsilon_r \nabla^2 V$$

where ε_0 is the vacuum permittivity (8.854×10^{-12} C V^{-1} m^{-1}) and ε_r is the relative dielectric constant. With this technique, the total charge density across the $LaAlO_3/SrTiO_3$ interface, including the interface-specific 2DEG, can be obtained directly from the electrostatic potential distribution retrieved by the phase information of the transmitted electron beam (Fig. 2).

This inline holography technique can be more easily implemented than the off-axis counterpart since it does not require the use of bi-prism nor reference wave transmitted through field-free vacuum nearby the region of interest. Furthermore, it provides a large field-of-view (> 1 μm), and high spatial resolution (~0.8 nm) and low detection limit of charge density (~10^{17} cm^{-3}). The combination of high-resolution TEM (or scanning TEM, STEM) imaging and inline holography provides a unique tool to link the atomic and electronic structure of materials to their microscopic properties, which allows the structural and electronic properties of the switchable interfaces to be probed in unprecedented detail.

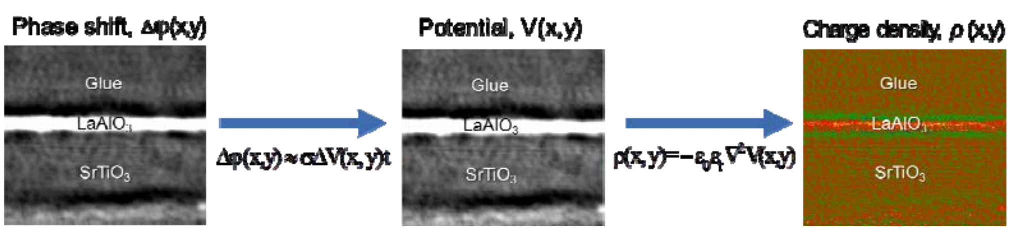

Figure 2. Conceptual representation of the processes to extract the internal electrostatic potential, *V (x, y)*, and the charge density, *ρ (x, y)*, distribution from the phase information of exit wavefunction.

Results and Discussion: Describe significant experimental and/or theoretical research advances or findings and their significance to the field and what work may be performed in the future as a follow on project. Fellow researchers will be interested to know what impact this research has on your particular field of science.

Direct imaging of 2DEGs by inline electron holography

We demonstrated that the inline electron holography can directly visualize the 2-DEG at the LAO/STO interface. At a very basic level, we find that the charge density map obtained from the 3 unit cell (u. c.) sample, which is thinner than the known critical thickness of 4 u. c., does not host significant charge density near the interface (Fig. 3a), and the 10 u. c. sample, greater than the known critical thickness exhibits the negative charges beneath the interface (Fig. 3b). Thus holography directly images the presence or absence of the interfacial 2DEG.

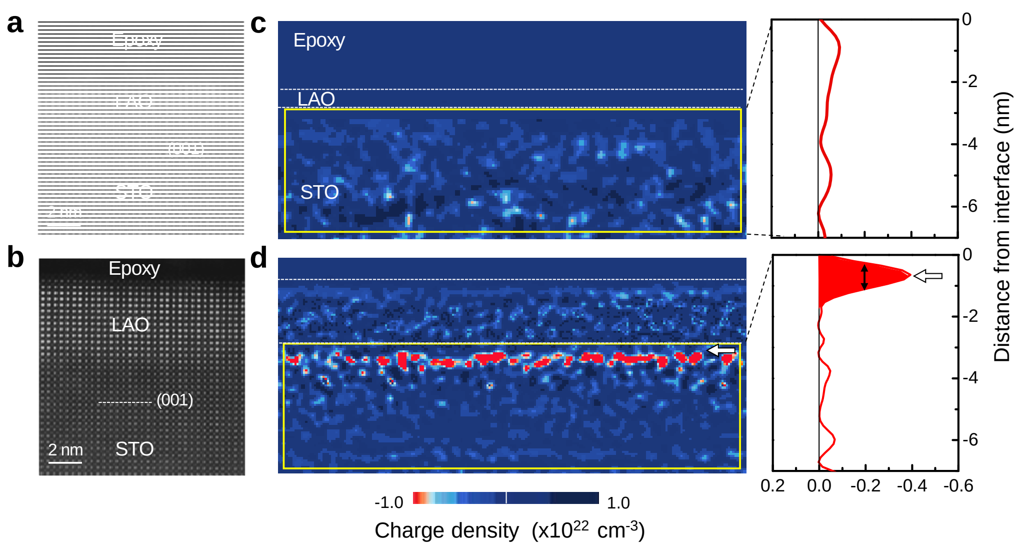

Figure 3. (a), (b) High-angle annular dark-field (HAADF) images of a 3-uc and 10-uc-thick $LaAlO_3$ film grown on $SrTiO_3$, oriented along the [001] direction, respectively. (c), (d) Corresponding 2-D charge density maps and profiles across the $LaAlO_3/SrTiO_3$ obtained by using inline electron holography. The white box in the charge density map represents the region of profile. The charge density map of the 3-uc $LaAlO_3/SrTiO_3$, with (001) orientation, does not show any significant charge density near the interface. In contrast, the interfacial 2-DEG was observed at 10-uc for (001)-interfaces. The 2-DEG is slightly off from the interface and confined in STO within about 1 nm at the (001)-interfaces.

But in addition, we resolved the spatial dependence of the charge density at the atomic level. The technique directly images electric potential, from which electric field is obtained directly. In order to extract a charge density of the 2-DEG from, one has to take account of the strong dependence of STO dielectric constant on electric field. This is essential, as the presence of the 2DEG leads to strong electric fields near the interface. The calibration of the dielectric constant using Landau theory yields a 2-DEG density close to the theoretically expected value corresponding to the transfer of 0.5 e per u. c. The intensity profile of the 2-DEG yields that the full width half maximum is 0.82 ± 0.34 nm, i.e. roughly two unit cell width (refer to the arrow in Fig. 3d). Our results also imply that the interfacial 2-DEG most likely arises from electrons transferred from oxygen vacancies near the LAO surface. By contrast, the charge density map of the 3 u. c. sample, which is below the critical thickness, does not show any significant charge density near the interface. In the absence of 2-DEG, the electric field in the 3 u.c. LAO film is partially compensated by the depolarization field induced by atomic displacement. Our results demonstrate the potential of inline electron holography for quantitative mapping of confined / modulated charge state at a hetero-interface or surface of oxide film.

List of Publications and Significant Collaborations that resulted from your AOARD supported project: In standard format showing authors, title, journal, issue, pages, and date, for each category list the following:

a) papers published in peer-reviewed journals,

b) papers published in peer-reviewed conference proceedings,

c) papers published in non-peer-reviewed journals and conference proceedings,

d) conference presentations without papers,

1. "Direct Probing of Two-Dimensional Electron Gas at $LaAlO_3/SrTiO_3$ Interface using Electron Holography", Kyung Song, Sangwoo Ryu, Hyungwoo Lee, Si-Young Choi, Tula R. Paudel, Christoph T. Koch, Mark S. Rzchowski, Evgeny Y. Tsymbal, Chang-Beom Eom and Sang Ho Oh, MRS Fall Meeting, Symposium N: Frontiers in Complex Oxides, November 30 – December 5, 2014, Boston, MA.

2. "Direct Probing of Two-Dimensional Electron Gas at $LaAlO_3/SrTiO_3$ Interface using Electron Holography", Kyung Song and Sang Ho Oh, 2014 Korean Ceramic Society Fall Meeting, October 16 -17, 2014, Yoosu, Korea.

e) manuscripts submitted but not yet published, and

1. "Global electronic reconstruction at the atomically smooth, polar (111)-oriented oxide interface" S. Ryu, C. W. Bark, T. Hernandez, T. R. Paudel, Hua. Zhou, D. D. Fong, Y. Zhang, J. Podkaminer, X. Q. Pan, E. Y. Tsymbal, M. S. Rzchowski, and C. B. Eom, submitted to *Nature Materials* (2015)
2. "Direct imaging of the electron liquid at oxide interfaces", Kyung Song, Sangwoo Ryu, Hyungwoo Lee, Si-Young Choi, Tula R. Paudel, Christoph T. Koch, Mark S. Rzchowski, Evgeny Y. Tsymbal, Chang-Beom Eom and Sang Ho Oh, *manuscript in preparation* (2015)

f) provide a list any interactions with industry or with Air Force Research Laboratory scientists or significant collaborations that resulted from this work.

We have developed research collaboration with Igor Altfeder and Andrey Voevodin at AFRL at Dayton, OH. They have been looking on a development of interfacial 2DEG using their UHV-STM with and without electrostatic gate voltage to determine the evolution of 2DEG charge density. This investigation is important for understanding oxide-based 2DEG field effect transistors and complimentary with this project. The Eom group at UW-Madison designed and provided symmetric n-type 2DEG bilayer heterostructures prepared by atomic layer engineering method. The AFRL group observed surface conductivity which is the indicator of 2DEG presence. We plan to extend these studies and correlate with the e-beam holography studies by the Oh group at POSTECH.

Dr. Andrey Voevodin invited us to the AFRL to give seminars and discuss research collaborations. We visited the AFRL on June 2, 2015 and gave seminar presentations on "Oxide Electronics" (Chang-Beom Eom) and "High-resolution mapping of orbital engineered two-dimensional electron gases at oxide interfaces" (Sang Ho Oh).